IN THE UNITED STATES PATENT AND TRADEMARK OFFICE
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In re the Application of: Akira YAMAGUCHI et al.
Application No.10/551,367
Filed: November 15, 2005

Group Art Unit: 1793
Examiner: Y. TAKEUCHI

For: ELECTRODE STEP DIFFERENCE ABSORBING PRINT PASTE AND METHOD OF PRODUCING ELECTRONIC DEVICE

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The Honorable Commissioner of Patents and Trademarks
United States Patent and Trademark Office
Washington, D. C. 20231

DECLARATION UNDER 37 CFR 1.132

Sir:

I, Toshio Sakurai, declare and state that:

1. In March 1994, I was graduated from Nihon University, Faculty of Humanities and Sciences and received a degree of Bachelor of Sciences from the same University. In March 1996, I was graduated from the master course of Tokyo Institute of Technology Faculty of Science, majoring in chemistry, and received a degree of Master of chemistry from the same University.

Since 1996, I have been an employee of TDK Corporation, and till the present time I have been engaged in research of mixing technique of ceramics , in research of materials of ceramics and development of the application.

2. I am familiar with the invention described in the specification of the above-identified application.

3. I carried out the following experiment. Details of my experiment are as follows.

Experiment (comparative)

(Production of Green Sheet, Release Layer Slurry, Adhesive Layer Slurry, & Internal Electrode Paste)

The green sheet, the release layer slurry, the adhesive layer slurry and the internal electrode paste were respectively prepared according to the method disclosed in the present specification, paragraphs [0121] to [0130].

Formation of Green Sheet and Transfer of Adhesive Layer and Electrode Layer

(Production of Electrode Level Difference Absorbing Print Paste)

The same ceramic powder and subcomponent additives were prepared as with the green sheet slurry to obtain the same compounding ratio. That is, 1.48 parts by weight of $(Ba_{0.6}Ca_{0.4})SiO_3$, 1.01 parts by weight of Y_2O_3, 0.72 wt% of $MgCO_3$, 0.13 wt% of Cr_2O_3 and 0.045 wt% of V_2O_5 were used with respect to 100 parts by weight of the $BaTiO_3$ powder (BT-02 made by Sakai Chemical Industry Co., Ltd.).

Ceramic powder and subcomponent additives (150g) was added with a dispersant of an ester based polymer (1.5g), an imidazoline based antistatic agent (0.6g), terpineol (50g) and diotycle phthalate as a plasticizer (5g) and mixed for 4 hours. Next, the mixed solution was added with 8% lacquer (8 wt% of polyvinyl butyral and 92 wt% of terpineol with respect to the entire lacquer) of BH6 (a polyvinyl butyral resin having a polymerization degree and a butyralation degree of 69 mol% ± 3%) made by Sekisui Chemical Co., Ltd. by an amount of 120g and mixed for 16 hours. After that, 0 to 60g of terpineol was added for viscosity adjustment to produce a paste.

As shown in Table 11, electrode level difference absorbing print pastes of sample numbers 1 to 8 were produced by changing a ceramic powder content (pigment concentration /wt%) with respect to the entire paste becomes 30 to 58 wt%.

Electrode level difference absorbing print pastes of sample numbers 10 to 17, 20 to 27, 30 to 37 and 40 to 47 were produced as with the above sample numbers 1 to 8 except for using a polyvinyl butyral resin having a polymerization degree of 1700, 2000, 2400 and 3000 respectively as a binder resin (see Tables 12 to 15).

Electrode level difference absorbing print pastes of sample numbers 50 to 54 were produced as with the above sample numbers 1 to 8 except for using a polyvinyl butyral resin having a polymerization degree of 2000 as a binder resin, and making a weight ratio (resin amount) of the binder resin with respect to 100 parts by weight of the ceramic powder to 2 to 10 parts by weight (see Table 16).

Electrode level difference absorbing print pastes of sample numbers 60 to 64 were produced as with the above sample numbers 1 to 8 except for using a polyvinyl butyral resin having a polymerization degree of 2400 as a binder resin, and changing a butyralation degree

thereof to be in a range of 77 to 63 mol% (see Table 17).

Electrode level difference absorbing print pastes of sample numbers 70 to 74 were produced as with the above sample numbers 1 to 8 except for using a polyvinyl acetal resin having a polymerization degree of 2400 as a binder resin, and changing an acetalization degree thereof to be in a range of 77 to 63 mol% (see Table 18).

Electrode level difference absorbing print pastes of sample numbers 80 to 86 were produced as with the above sample numbers 1 to 8 except for using a polyvinyl butyral resin having a polymerization degree of 2000 as a binder resin, and making dioctyl phthalate as a plasticizer contained at a ratio of 0 to 150 parts by weight with respect to 100 parts by weight of the binder resin (see Table 19).

Electrode level difference absorbing print pastes of sample numbers 90 to 94 were produced as with the above sample numbers 1 to 8 except for using a polyvinyl butyral resin having a polymerization degree of 2000 as a binder resin and using any one of polyethylene glycol (a hygroscopic polymer), polyalkylene glycol derivative based surfactant (amphoteric surfactant), carboxylic acid amidine salt based surfactant (amphoteric surfactant), and imidazoline based surfactant (amphoteric surfactant) as an antistatic agent, or not adding any antistatic agent (see Table 20).

(Formation of Green Sheet and Transfer of Adhesive Layer and Electrode Layer)

Further, the green sheet was formed, and the adhesive layer and the electrode layer were transferred according to the specification, paragraphs [0140] to [0145].

For all of the above samples, "hanging of paste", "stacking property (stacking precision)" and "sheet erosion" were examined as well as other properties such as viscosity of the pastes and the minimum possible printing thicknesses by the printing method (a print material thickness).

Note that the "hanging of paste" column indicates to what extent the paste hangs over the electrode from the edges. This "hanging paste" phenomenon results when a paste has a viscosity that is too low. In other words, a paste with a low viscosity is unstable, loose, and will not keep its intended figure and, for example, the paste will hang over the edges of the electrode as described in the specification, paragraph [0136]. Additionally, "stacking property (stacking precision)" and "sheet erosion" also are deteriorated when a paste has a viscosity that is too low.

From the results of the above experiment, and based on my knowledge and experience on production of electrode level difference absorbing print paste, I conclude that:

It is important to control the viscosity of the electrode level difference absorbing print paste within the claimed range.

The undersigned declares further that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Respectfully submitted,

This 26 th day of March 2009

Toshio Sakurai

Toshio Sakurai

Table 11

	Polymerization Degree	Resin Amt [php]	Pigment Conc. [wt%]	Butyralation Degree [mol%]	Acetalization Degree [mol%]	Plasticizer Amt [php]	Antistatic Agent Kind	Viscosity [Pa·s] (at 8[1/s])	Print Thickness [μm]	Hanging of Paste	Stacking Property (Stacking Precision)	Sheet Erosion	Release Property of Sheet
Sample 1	1450	6	30	69		50	Imidazolines	4	0.7	No	Good (≤ 50 μm)	Little	Easy
Sample 2	1450	↓	34	↓		↓	↓	4	0.8	↓	↓	↓	↓
Sample 3	1450	↓	38	↓		↓	↓	5	1.0	↓	↓	↓	↓
Sample 4	1450	↓	42	↓		↓	↓	7	1.2	↓	↓	↓	↓
Sample 5	1450	↓	46	↓		↓	↓	11	1.6	↓	↓	↓	↓
Sample 6	1450	↓	50	↓		↓	↓	18	1.9	↓	↓	↓	↓
Sample 7	1450	↓	54	↓		↓	↓	30	2.2	↓	↓	↓	↓
Sample 8	1450	↓	58	↓		↓	↓	52					

Table 12

	Polymerization	Resin Amt [php]	Pigment Conc. [wt%]	Butyralation Degree [mol%]	Acetalization Degree [mol%]	Plasticizer Amt [php]	Antistatic Agent Kind	Viscosity [Pa·s] (at 8[1/s])	Print Thickness [μm]	Hanging of Paste	Stacking Property (Stacking Precision)	Sheet Erosion	Release Property of Sheet
Sample 10	1700	6	30	69		50	Imidazolines	1	0.7	Yes	Bad (≥ 100 μm)	Yes	Hard
Sample 11	1700	↓	34	↓		↓	↓	3	0.8	↓	↓	↓	↓
Sample 12	1700	↓	38	↓		↓	↓	5	1.0	No	Good (≤ 50 μm)	Little	Easy
Sample 13	1700	↓	42	↓		↓	↓	9	1.2	↓	↓	↓	↓
Sample 14	1700	↓	46	↓		↓	↓	17	1.6	↓	↓	↓	↓
Sample 15	1700	↓	50	↓		↓	↓	29	1.9	↓	↓	↓	↓
Sample 16	1700	↓	54	↓		↓	↓	45					
Sample 17	1700	↓	58	↓		↓	↓	70					

Table 13

	Polymerization	Resin Amt [php]	Pigment Conc. [wt%]	Butyralation Degree [mol%]	Acetalization Degree [mol%]	Plasticizer Amt [php]	Antistatic Agent Kind	Viscosity [Pa·s] (at 8[1/s])	Print Thickness [μm]	Hanging of Paste	Stacking Property (Stacking Precision)	Sheet Erosion	Release Property of Sheet
Sample 20	2000	6	30	69		50	Imidazolines	1	0.7	Yes	Bad (≥ 100 μm)	Yes	Hard
Sample 21	2000	↓	34	↓		↓	↓	4	0.7	No	Good (≤ 50 μm)	Little	Easy
Sample 22	2000	↓	38	↓		↓	↓	6	1.0	No	Good (≤ 50 μm)	Little	Easy
Sample 23	2000	↓	42	↓		↓	↓	11	1.2	↓	↓	↓	↓
Sample 24	2000	↓	46	↓		↓	↓	20	1.6	↓	↓	↓	↓
Sample 25	2000	↓	50	↓		↓	↓	37					
Sample 26	2000	↓	54	↓		↓	↓	64					
Sample 27	2000	↓	58	↓		↓	↓	90					

Table 14

	Polymerization	Resin Amt [php]	Pigment Conc. [wt%]	Butyralation Degree [mol%]	Acetalization Degree [mol%]	Plasticizer Amt [php]	Antistatic Agent Kind	Viscosity [Pa·s] (at 8[1/s])	Print Thickness [μm]	Hanging of Paste	Stacking Property (Stacking Precision)	Sheet Erosion	Release Property of Sheet
Sample 30	2400	6	30	69		50	Imidazolines	2	0.7	Yes	Bad (≥ 100 μm)	Yes	Hard
Sample 31	2400	↓	34	↓		↓	↓	5	0.7	No	Good (≤ 50 μm)	Little	Easy
Sample 32	2400	↓	38	↓		↓	↓	10	1.0	No	Good (≤ 50 μm)	Little	Easy
Sample 33	2400	↓	42	↓		↓	↓	16	1.2	↓	↓	↓	↓
Sample 34	2400	↓	46	↓		↓	↓	31					
Sample 35	2400	↓	50	↓		↓	↓	47					
Sample 36	2400	↓	54	↓		↓	↓	77					
Sample 37	2400	↓	58	↓		↓	↓	115					

Table 15

	Polymerization	Resin Amt [php]	Pigment Conc. [wt%]	Butyralation Degree [mol%]	Acetalization Degree [mol%]	Plasticizer Amt [php]	Antistatic Agent Kind	Viscosity [Pa·s] (at 8[1/s])	Print Thickness [μm]	Hanging of Paste	Stacking Property (Stacking Precision)	Sheet Erosion	Release Property of Sheet
Sample 40	3000	6	30	69		50	Imidazolines	4	0.5	Yes	Bad (≥ 100 μm)	Yes	Hard
Sample 41	3000	↓	34	↓		↓	↓	7	0.8	No	Good (≤ 50 μm)	Little	Easy
Sample 42	3000	↓	38	↓		↓	↓	15	1.1	No	Good (≤ 50 μm)	Little	Easy
Sample 43	3000	↓	42	↓		↓	↓	30					
Sample 44	3000	↓	46	↓		↓	↓	50					
Sample 45	3000	↓	50	↓		↓	↓	84					
Sample 46	3000	↓	54	↓		↓	↓	131					
Sample 47	3000	↓	58	↓		↓	↓	200					

Table 16

	Polymerization	Resin Amt [php]	Pigment Conc. [wt%]	Butyralation Degree [mol%]	Acetalization Degree [mol%]	Plasticizer Amt [php]	Antistatic Agent Kind	Viscosity [Pa·s] (at 8[1/s])	Print Thickness [μm]	Hanging of Paste	Stacking Property (Stacking Precision)	Sheet Erosion	Release Property of Sheet	Film Density [g/cm^3]
Sample 50	2000	2	42	69		50	Imidazolines	2	1.0	Yes	Bad (≥100 μm)	Yes	Hard	3.8
Sample 51	↓	4	↓	↓		↓	↓	4	1.1	No	Good (≤ 50 μm)	Little	Easy	3.8
Sample 52	↓	6	↓	↓		↓	↓	11	1.2	No	Good (≤ 50 μm)	Little	Easy	3.6
Sample 53	↓	8	↓	↓		↓	↓	20	1.3	No	Good (≤ 50 μm)	Little	Easy	3.4
Sample 54	↓	10	↓	↓		↓	↓	35						

Table 17

	Polymerization	Resin Amt [php]	Pigment Conc. [wt%]	Butyralation Degree [mol%]	Acetalization Degree [mol%]	Plasticizer Amt [php]	Antistatic Agent Kind	Viscosity [Pa·s] (at 8[1/s])	Print Thickness [μm]	Hanging of Paste	Stacking Property (Stacking Precision)	Sheet Erosion	Release Property of Sheet	Surface Roughness Ra[μm]
Sample 60	2400	6	42	77		50	Imidazolines	3	1.2	Yes	Bad (≥ 100 μm)	Yes	Hard	0.55
Sample 61	↓	↓	↓	74		↓	↓	8	1.3	No	Good (≤ 50 μm)	Little	Easy	0.59
Sample 62	↓	↓	↓	69		↓	↓	16	1.3	No	Good (≤ 50 μm)	Little	Easy	0.62
Sample 63	↓	↓	↓	66		↓	↓	20	1.4	No	Good (≤ 50 μm)	Little	Easy	0.91
Sample 64	↓	↓	↓	63		↓	↓	33						

Table 18

	Polymerization	Resin Amt [php]	Pigment Conc. [wt%]	Butyralation Degree [mol%]	Acetalization Degree [mol%]	Plasticizer Amt [php]	Antistatic Agent Kind	Viscosity [Pa·s] (at 8[1/s])	Print Thickness [μm]	Hanging of Paste	Stacking Property (Stacking Precision)	Sheet Erosion	Release Property of Sheet	Surface Roughness Ra[μm]
Sample 70	2400	6	42		77	50	Imidazolines	36						
Sample 71	↓	↓	↓		74	↓	↓	28	1.4	No	Good (≤ 50 μm)	Little	Easy	0.59
Sample 72	↓	↓	↓		69	↓	↓	23	1.4	No	Good (≤ 50 μm)	Little	Easy	0.62
Sample 73	↓	↓	↓		66	↓	↓	14	1.3	No	Good (≤ 50 μm)	Little	Easy	0.91
Sample 74	↓	↓	↓		63	↓	↓	3	1.3	Yes	Bad (≥ 100 μm)	Yes	Hard	0.60

Table 19

	Polymerization	Resin Amt [php]	Pigment Conc. [wt%]	Butyralation Degree [mol%]	Acetalization Degree [mol%]	Plasticizer Amt [php]	Antistatic Agent Kind	Viscosity [Pa·s] (at 8[1/s])	Print Thickness [μm]	Hanging of Paste	Stacking Property (Stacking Precision)	Sheet Erosion	Release Property of Sheet	PET Release Force [mN/cm]
Sample 80	2000	6	42	69		0	Imidazolines	12	1.1	No		Little	Hard	35
Sample 81	↓	↓	↓	↓		10	↓	12	1.1	No	Good (≤ 50 μm)	Little	Easy	25
Sample 82	↓	↓	↓	↓		30	↓	12	1.2	No	Good (≤ 50 μm)	Little	Easy	20
Sample 83	↓	↓	↓	↓		50	↓	12	1.2	No	Good (≤ 50 μm)	Little	Easy	16
Sample 84	↓	↓	↓	↓		80	↓	12	1.2	No	Good (≤ 50 μm)	Little	Easy	14.00
Sample 85	↓	↓	↓	↓		100	↓	11	1.3	No	Good (≤ 50 μm)	Little	Easy	10.00
Sample 86	↓	↓	↓	↓		150	↓	10	1.3	Yes				Unmeasurable

Table 20

	Polymerization	Resin Amt [php]	Pigment Conc. [wt%]	Butyralation Degree [mol%]	Acetalization Degree [mol%]	Plasticizer Amt [php]	Antistatic Agent Kind	Viscosity [Pa·s] (at 8[1/s])	Print Thickness [μm]	Hanging of Paste	Stacking Property (Stacking Precision)	Sheet Erosion	Release Property of Sheet	Static Electrocity Amt [kV]
Sample 90	2000	6	42	69		50	*1	12	1.1	No	Good (≤ 50 μm)	Little	Easy	12
Sample 91	↓	↓	↓	↓		↓	*2	12	1.2	No	Good (≤ 50 μm)	Little	Easy	11
Sample 92	↓	↓	↓	↓		↓	*3	12	1.2	No	Good (≤ 50 μm)	Little	Easy	13
Sample 93	↓	↓	↓	↓		↓	*4	12	1.2	No	Good (≤ 30 μm)	Little	Easiest	4
Sample 94	↓	↓	↓	↓		↓	None	12	1.2	No	Bad (≥ 100 μm)	Little	Hard	36.00

*1 polyethylene glycol
*2 polyalkylene glycol derivative based surfactant
*3 carboxylic acid amidine salt based surfactant
*4 imidazoline based surfactant